state	(MeV)	H	He	Li	B	С	0	F	Al	Si	P	S	Cl	Sc	Cr	Fe	Ni	Cu	Br	Ag	Ι	Ta	Au
14	90												${\color{black} \big }$		U	TT	4 <mark>¢</mark> -	93,	202	24	$\left(\right)$		\bigwedge
13	84												Δ	Δ			0	\underline{V}	\bigwedge	\bigwedge	0		Δ
12	78													Δ	0	${\color{black} \big }$	0	0	0	0	0		0
11	72				A	N	N	U	A L	. F	RE		OF	R.L.	2	02	23	0	0	0	0		0
10	66									0	Δ	0	0	Δ	0	0	0	0	0	0	0		0
9	60								0	0	0	0	0	Δ	0	0	0	0	0	0	0	Δ	0
8	54						0	0			0		0				0	0	0	0	0	${\color{black} \big }$	0
7	48											ES f To					0	0	0	0	0		
6	42					0	0	0						Ja		0	0	0	0	0			
5	36				0	0	0	0	0	0	0	0	0			0	0	0	0	0			
4	30				0	0	0	0	0	0	0	0	0			0	0	0	0				
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2	18		0	0	0	0	0		\bigwedge		0	0	0										
1	12	0	0	0																			

UTTAC ANNUAL REPORT 2023

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UTTAC-93, 2024

Editors : Tetsuaki Moriguchi, Kimikazu Sasa, Yoshihiro Yamato, Masumi Matsumura, Masao Sataka, Hiroshi Naramoto, Eiji Kita, and Hiroshi Kudo (editor-in-chief)

UTTAC ANNUAL REPORT is a series of issues, which describes annual research activities at Tandem Accelerator Complex, Research Facility Center for Science and Technology, University of Tsukuba.

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Cover: Table of typical ion beam intensities shown with corresponding marks, which is updated annually as a guide for users of the 6MV tandem accelerator at UTTAC. —*from Report 1.3.*

PREFACE

This annual report covers research and development (R&D) carried out at University of Tsukuba Tandem Accelerator Complex (UTTAC) during the fiscal year 2023 (1 April 2023 \sim 31 March 2024). The topics include not only accelerator-based R&D using the 6MV tandem and 1MV Tandetron accelerators, but also radioisotope-based R&D employing positron annihilation spectroscopy and Mössbauer spectroscopy.

Additionally, UTTAC was reorganized as one of the divisions under the Center for Research in Radiation, Isotopes, and Earth System Sciences (CRiES), effective on April 1, 2023.

September 1, 2024 Editorial Board



Evacuation drill in fall 2023 (in front of the entrance of UTTAC)

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1.

ACCELERATOR OPERATION AND RELATED STUDIES



Engineers of UTTAC working for maintenance of the 6MV tandem accelerator.

1.1 Accelerator operation 2023

T. Moriguchi, S. Ishii, T. Takahashi, Y. Yamato, T. Yoshida, M. Matsumura, T. Nakazawa, K. Sasa

University of Tsukuba Tandem Accelerator Complex (UTTAC) is promoting the maintenance and operation of the tandem accelerator facility consisting of the 6MV Pelletron tandem accelerator and the 1MV Tandetron accelerator for cooperative researches both inside and outside the University of Tsukuba. In October 2023, during the annual inspection of power supplies on campus, 400V AC was incorrectly applied to the 200V AC power line of UTTAC. As a result, the inverter power supplies for the chain motors of the 6MV tandem accelerator and the relay switches in the distribution board failed, hence all the scheduled beam time was suspended for about two months. The damaged equipment was fully restored in late March 2024.

1MV Tandetron accelerator

The 1MV Tandetron accelerator has 2 negative ion sources and 4 beamlines. The operating time and the experimental beam time of the 1 MV Tandetron accelerator were 908 and 250 hours, respectively, during the total service time in fiscal 2023. The total number of operation days was 50, and the total of 26 research programs was carried out. Figures 1 and 2 show classification of the accelerated ions and of the experimental purposes, respectively. At the end of March 2024, the number of approved research programs was 3 for on-campus and 7 for off-campus. The terminal voltage instability reported last year [1] was also observed frequently in FY2023, and the investigation of this issue is now underway.



Fig. 1. Accelerated ions from the 1MV Tandetron accelerator in FY2023.



Fig. 2. Purposes of use of the 1MV Tandetron accelerator in FY2023.

6MV Pelletron tandem accelerator

The 6MV Pelletron tandem accelerator has 5 negative ion sources and 12 beamlines. The operation time and the experimental beam time of the 6MV Pelletron tandem accelerator were 1,022 and 832 hours, respectively, during the total service time in fiscal 2023. The total number of operation days was 96, and the total of 57 research programs was carried out. Figure 3 shows the beam time histogram with respect to the terminal voltage. Figures 4 and 5 show classification of the accelerated ions and of the experimental

purposes, respectively. At the end of March 2024, the number of approved research programs was 8 for on-campus and 5 for off-campus.



Fig. 3. Beam time histogram as a function of the terminal voltage for the 6MV Pelletron tandem accelerator in FY2023.



Fig. 4. Accelerated ions from the 6MV Pelletron tandem accelerator in FY2023.



Fig. 5. Purposes of use of the 6MV Pelletron tandem accelerator in FY2023.

Reference

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1.2 Production of a faint ⁵⁸Ni beam

T. Moriguchi, K. Sasa, Y. Yamato, T. Yoshida, S. Ishii, T. Takahashi, M. Matsumura, T. Nakazawa

For research and development of particle detectors, a faint beam is necessary to evaluate their performances without deterioration. In the last FY, we developed a faint beam of 12 MeV H^+ which was transported to the A7 course in the experimental room [1]. In FY2023, we transported a faint ⁵⁸Ni beam to the A7 course, as reported below.

A ⁵⁸Ni beam from the 6MV tandem accelerator cannot be extracted to the air for measurement of the count rate, unlike 12 MeV protons, since the Ni beam stops in a thin vaccum-seal window. Therefore, we installed a plastic scintillation counter (PL) at the scattering chamber in the A7 course. The disc-shaped plastic scintillator (Saint-Gobain BC400, $\phi 1$ in., 3 mm thick) was mounted with a photomultiplier tube (HAMAMATSU PHOTONICS K.K., R9880U-210) using a transparent gel (Shin-Etsu Chemical Co., Ltd., KE-1051J-A/B). The diameter of the plastic scintillator is almost the same as that of the effective area of a standard Faraday cup in the beam line. As shown in Fig. 1 (a), the PL was attached with the target ladder in the scattering chamber along with an alumina fluorescent plate (Desmarquest Co., AF995R) and a ZnS:Ag phosphor applied to a clear polyester plastic sheet (ELJEN TECHNOLOGY, EJ440).



Fig. 1. (a) A plastic scintillation counter, an alumina plate, and a ZnS:Ag sheet which are attached with the target ladder in the A7 course. (b) The beam spot on a ZnS:Ag sheet.

To produce a faint beam, a beam of 84 MeV 58 Ni¹³⁺ with a full intensity of ~0.2 nA, which was measured by the A7 Faraday cup, was firstly transported to the A7 course. While maintaining the full intensity, the beam spot was adjusted by using the ZnS:Ag sheet because the alumina plate was much less luminous (Fig. 1 (b)). Afterwards, we inserted an attenuator upstream of the accelerator, and closed the low-energy beam slits (LE-slit) to the lower limit, indicating the 1.0-mm squared aperture. At this time, the beam count rates of ~100 cps were measured by the PL at the A7 course. For attenuation of the 58 Ni intensity, defocussing of the beam without activating the quadrupole magnet was not needed unlike faint protons because the full intensity of ${}^{58}Ni^{13+}$ was low. By varying the aperture of the LE-slit, the count rate was able to be optimized as requested. The faint ${}^{58}Ni^{+13}$ beam was provided for the performance test of position-sensitive detectors [2]. It is notable that in the similar trial the beam intensity of ${}^{58}Ni^{14+}$ was too low to be transported [3].

The present development was conducted as part of a course of Target Oriented Group Study offered by the College of Physics, and we thank K. Matsuda, who was a student of this course, for the preparation of the PL.

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[1] T. Moriguchi et al., UTTAC Annual Report 2022, UTTAC-92 (2023) 5.

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1.3 Search for the highest possible energy of the ions produced by the 6 MV tandem accelerator

M. Sataka, T. Takahashi, H. Naramoto, H. Kudo, K. Sasa

Acceleration tests of ion beams obtained from the 6MV tandem accelerator at UTTAC have been continued [1]. High-energy heavy ion beams are required for ion irradiation experiment for materials science such as ion beam modification of materials [2,3]. High-energy ion beams of very weak intensities are indispensable for radiation resistance tests of semiconductors used in space [4].

We have measured beam currents of the accelerated ions as a function of the ion charge at the acceleration voltage from 3 to 6 MV. In this fiscal year, data of P ions were newly obtained, while the previous Si data were updated. Figure 1 shows the beam current distributions of P ions as a function of the ion charge at the maximum terminal voltage of 6 MV. A carbon stripper foil of approximately 5 μ g/cm² thickness was used at the 6 MV terminal.

In this accelerator control system, the minimum measurable current of the Faraday cup is 0.1 nA. However, it is required to measure lower currents to search for the maximum ion energy. In the test measurement, the bending magnets, lenses and steerers were preset to the estimated values, thereby leading the ion beam to the end Faraday cup with a picoammeter at the experimental apparatus. The beam operation time was not significantly different from that for high current ions. For lower current ion beams, a particle detector was used instead of a picoammeter.



Fig. 1. Ion beam current distribution for P ions at the terminal voltage of 6 MV. A foil stripper was used in the high voltage terminal.

Table 1 shows the ion charges and maximum energies obtained by injection of negative atomic ions at the highest terminal voltage of 6 MV. These data were accumulated from 2016 to 2023.

Table 1. Summary of the acceleration tests from 2016 to 2023 for injection of negative atomic ions at the highest terminal voltage of 6 MV. The measured beam intensities are shown by the symbols: $O: \ge 1nA, \Delta: \le 1nA$. Shown in red are those obtained by using foil strippers.

charge	energy																						
state	(MeV)	Н	He	Li	В	С	0	F	Al	Si	Р	S	Cl	Sc	Cr	Fe	Ni	Cu	Br	Ag	Ι	Та	Au
14	90												\triangle		\triangle		\triangle		Δ	\triangle	0		\triangle
13	84											\triangle	\triangle	\triangle	\triangle		0	\triangle	\triangle	\triangle	0		\triangle
12	78											\triangle	\triangle	\triangle	0	Δ	0	0	0	0	0		0
11	72											\triangle	Δ	\triangle	0	0	0	0	0	0	0		0
10	66									0	\triangle	0	0	\triangle	0	0	0	0	0	0	0		0
9	60								0	0	0	0	0	\triangle	0	0	0	0	0	0	0	\bigtriangleup	0
8	54						0	0	0	0	0	0	0	Δ	0	0	0	0	0	0	0	\triangle	0
7	48						0	0	0	0	0	0	0	Δ	0	0	0	0	0	0	0		
6	42					0	0	0	0	0	0	0	0			0	0	0	0	0			
5	36				0	0	0	0	0	0	0	0	0			0	0	0	0	0			
4	30				0	0	0	0	0	0	0	0	0			0	0	0	0				
3	24			0	0	0	0	0	0	0	0	0	0			0	0						
2	18		0	0	0	0	0	0	\triangle		0	0	0										
1	12	0	0	0																			

Some ion beams are formed from negative molecular ions (e.g., nitrogen and Ca ion beams), rather than from negative atomic ions. In these cases, the energies of the accelerated ions are not given in Table 1, as those cannot be determined solely from the terminal voltage and the ion charge after acceleration. The data of molecular ions (N, Al, Ca, Sc, Ti, Mo, W) accumulated from 2016 to 2022 were presented in the previous report [1].

References

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- [4] H. Kukita et al., in this annual report 5.3.

2.

NUCLEAR AND COLLISION-BASED RESEARCH



Development of a high-sensitivity X-ray detector.

2.1 Beam test of position-sensitive detectors for the RIKEN Rare-RI Ring facility

T. Yamaguchi^{1,2}, K. Sasaki², K. Takiura², Y. Kikuchi², S. Nishizawa², N. Tomioka², K. Watanabe², T. Suzuki², T. Moriguchi, A. Ozawa, A. Yano

We have been developing the storage ring facility called Rare-RI Ring (R3) [1] at the RI Beam Factory in RIKEN for precision mass spectrometry of exotic nuclei. Rare-RI Ring is coupled with the RIBF cyclotron facility and hence individually stores a single ion of interest at a time, which is in-flight selected at the fragment separator BigRIPS. To realize such a new injection scheme, a long injection beam line is equipped, where many beam diagnostics detectors need to be installed. They should be easily operated, maintained and are required to equip with a low cost. More important is to confirm the ion-optical condition, emittance and dispersion matching at the injection kicker position. Therefore, a compact design that fits into the gap in the kicker chamber is required. For this purpose, we have developed several shaped plastic scintillation detectors for position detection, and tested with swift heavy ions at the Heavy Ion Medical Accelerator in Chiba (HIMAC) facility [2] and with a 12-MeV proton beam at the 6-MV tandem accelerator in University of Tsukuba.

In the previous annual report in 2021 [3], we reported an one-dimentional position-sensitive detector. The detector consists of a simple plastic scintillator plate (100 × 100 mm², 3 mm thick) and three bundled wavelength shifters, WLS (each 1 × 1 mm², Kuraray Y-11), which is an easy and cost effective solution. The WLS are connected at the up and down side of the scintillator plate, and the photons from WLS are read out by four photo sensors at four corners: left-up (LU), left-down (LD), right-up (RU), and right-down (RD). A photo of the detector is shown in Fig. 1 of ref. [3]. For a 200-MeV/u Kr beam at the HIMAC facility, a position resolution of 1.2 mm (σ) in the y-direction was obtained from the pulse height ratio which is defined as $E_y = (Q_{LU} - Q_{LD} + Q_{RU} - Q_{RD})/(Q_{LU} + Q_{LD} + Q_{RU} + Q_{RD})$. Here, Q_i means an integrated charge measured with a charge-sensitive analog-to-digital converter, and the subscript *i* means each position of the photo sensors as described above. A linear relation in a wide range has been achieved.

Through several experiments done so far, it turns out that the position resolution is inversely proportional to the square root of the energy deposit, as shown in Fig. 1(a). To confirm this character, we employed a Ni¹³⁺ beam with an energy of 84 MeV this time [4]. All the detector setups were installed in a vacuum chamber connected at the end of the A7 beam course. Although the expected energy deposit ($\Delta E^{-1/2} \approx 0.11 \text{ MeV}^{-1/2}$) should be around the middle of the fitting line in Fig. 1(a), the observed pulse heights were unexpectedly small due to a short range in plastic, $\approx 20 \ \mu$ m, and thus possible photon transport deteriorated. However, the result of position resolution, $\approx 15 \text{ mm}$, consistently matches the straight line, meaning an energy deposit of 4-5 MeV equivalent to the offline test with a standard α -source ²⁴¹Am.

To further upgrade this detector, the present position-sensitive plastic scintillation detector is coupled with a scintillating fiber detector [4]. On the one hand, the present detector has a limited position resolution depending on the energy deposit of beam, whereas it is compact and simple readout. On the other hand, the scintillating fiber detector has an excellent resolution, while fiber readout is generally complicated. Thus, to

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Fig. 1. (a) Position resolution as a function of the inverse square root of energy deposit into the present detector. (b) Proton beam distribution measured by the prototype scintillating-fiber detector.

combine both features is beneficial.

A prototype scintillating fiber detector, which consists of 64 scintillating fibers with a square cross section of $1 \times 1 \text{ mm}^2$, was built and successfully tested by using a 12-MeV proton beam. The light output is read out from both ends of the fibers with Multi-Pixel Photon Counters (Hamamatsu S13360-6075CS). To reduce the number of readout channels, in one end, successive 8 fibers out of 64 fibers are connected together into a MPPC, and at the other end every 8 fibers out of 64 fibers are connected in the same manner. In total 16 MPPC are used. By combining the hit patterns of MPPC at both ends, one can reconstruct the hit position out of the 64 fibers, that is the beam position distribution. The present readout method is based on a previous work [5]. A typical result of 12-MeV proton beam distribution is shown in Fig. 1(b).

We have recently built new detectors with 100 fibers of 1-mm diameter and with 300 fibers of 0.5-mm diameter, and are ready for beam test. The developments are ongoing.

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2.2 The nuclear polarization of ²⁹P in semiconductor Si stoppers

A. Yano, A. Ozawa, T. Moriguchi, Y. Yamato, K. Tomita, N. Kaname, M. Hayashi, A. Moriyama, H. Zhang,T. Matsumoto

It is known that nuclear polarization depends on beam energies and stopper materials. Thus, as a prior step of the study of ³⁰P nuclear polarization, we investigated the nuclear polarization of ²⁹P with various types of semiconductor Si stoppers and various beam energies. This approach is reasonable because all P isotopes should have the same magnetic dipole interactions in stoppers.

We irradiated the Si targets with polarized proton and deuteron beams. Then, polarized ²⁹P was generated via polarization transfer reaction. Because we used thick targets, production targets and stopper materials are the same in our experiments. Experimental setup is similar to the previous experiment [1].

Typical β -NMR spectrum is shown in Fig. 1. The horizontal axis represents the applied RF frequency, while the vertical axis represents 4AP, where A is the asymmetry parameter of ²⁹P and P is the polarization of ²⁹P. Note that 4AP is not corrected for solid angles in detectors and effects of contaminants. Here, we define the β -NMR effect as the difference between 4AP for RF off and at 2.65 MHz which corresponds to the resonance in Fig. 1. Fig. 2 shows beam energy dependence of the nuclear polarization of ²⁹P. Vertical axis indicates calculated polarization, \tilde{P} , from the β -NMR effects. Polarized deuteron beams induce ~6% polarization, and it is almost constant. In contrast, polarized proton beams induce less than 2% polarization, and the lower the beam energy, the smaller the polarization.



Fig. 1. Typical β -NMR spectrum. Experimental conditions are written in bottom left.



Fig. 2. Beam energy dependence of the nuclear polarization of ²⁹P.

Regarding the stopper material dependence, obvious differences were observed in the previous experiment [2] for four types of Si stoppers with polarized proton beams. In this study, we conducted similar measurements with polarized deuteron beams. As a result, we obtained a more significant dependence with polarized deuteron beams compared to polarized proton beams, as shown in Fig. 3. The label high (low) indicates the high (low) –impedance. To clarify this dependence, we conducted spin relaxation measurements as well. We show the experimental data and the fitted curves in Fig. 4. UD ratio on the vertical axis indicates the ratio of the number of spin–up ²⁹P to the number of spin–down ²⁹P. Figure 4(a) shows the Si (n-type, low) data, while Fig. 4(b) shows the Si (p-type, low) data. To determine spin relaxation times, we fitted the experimental data with equations below. Equation 1 is for Fig. 4(a), and Eq. 2 is for Fig. 4(b) where a1, b1, a2, b2, c1, a, b, and c are fitting parameters.

$$fit1(x) = a1 \cdot \exp\left(-\frac{x}{b1}\right) + a2 \cdot \exp\left(-\frac{x}{b2}\right) + c1 \tag{1}$$

$$fit(x) = a \cdot \exp\left(-\frac{x}{b}\right) + c \tag{2}$$

Figures 4(a) and 4(b) have a common component with a spin relaxation time of ~9 s corresponding to b1 and b. Additionally, Fig. 4(a) has a component with the opposite spin direction and a long spin relaxation time of ~18 s corresponding to b2. The cause of this opposite component could be the contaminants polarization or the trapping of ²⁹P in at least two sites of Si (n-type, low) unlike Si (p-type, low). To understand the reason of this long relaxation time, we will measure the resonance of ²⁹P precisely and will determine its width.



Fig. 3. Target dependence of the β -NMR effects. The solid triangles correspond to 8 MeV, and the open triangle corresponds to 10 MeV. For comparison, the data of polarized proton beam with beam energy of 12 MeV are plotted with circles.



Fig. 4. (a) Spectrum of the spin relaxation with Si (n-type, low), and (b) with Si (p-type, low).

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2.3 Determining the composition of mixed molecular ion beams in the 1 MV Tandetron accelerator using Rutherford backscattering

Y. Yano, U. Ozeki, K. Minami, T. Nakazawa, S. Ishii, K. Sasa, S. Tomita

The acceleration of molecular ions using tandem accelerators is of great interest in many research fields. However, the accelerated molecular ion beam can often be a mixture of intact molecular ions with the same mass-to-charge ratio. The ability to determine the composition of these mixed molecular ion beams is not only crucial but also has significant practical implications, as it allows us to optimize the beam for specific applications.

This study presents a novel application of the Rutherford backscattering method to quantify individual components in mixed molecular ion beams accelerated by the 1 MV Tandetron accelerator at UTTAC. From the analysis of the Rutherford backscattering spectra of the accelerated ions, the proportions of the molecular and atomic components could be determined, providing the composition of the mixed beam.

The experiments were conducted at the 1 MV Tandetron accelerator at UTTAC. We selected a mixed ion beam with m/q = 13, comprising the molecular ion ${}^{12}\text{CH}^+$ and the atomic ion ${}^{13}\text{C}^+$, as a representative example. These ions were initially generated by sputtering graphite with Cs ions. To estimate the pre-acceleration proportions of ${}^{12}\text{CH}^+$ and ${}^{13}\text{C}^+$, we measured the beam currents of m/q = 12 and 13 using a Faraday cup located upstream of the accelerator. Assuming the natural abundance for the intensity ratio of ${}^{12}\text{C}^+$ and ${}^{13}\text{C}^+$, we estimated the current of ${}^{13}\text{C}^+$ in the beam m/q = 13 to be 11%.

Rutherford backscattering was employed to determine the composition of the accelerated mixed molecular ion beam. The beam of m/q = 13 ions was accelerated to 120 keV/u, and incident on a thin 57.8 nm Au film target evaporated on highly oriented pyrolytic graphite (HOPG). The energy spectra of the backscattered ions were measured using a Passivated Implanted Planar Silicon detector (PIPS) placed at 150° relative to the incident beam.

Rutherford backscattering spectra of the accelerated m/q = 13 ion beam showed the superposition of the ¹²C⁺ and ¹³C⁺ spectra, originating from the molecular ion ¹²CH⁺ and the atomic ion ¹³C⁺, respectively (Fig. 1). Simulations of the Rutherford backscattering of ¹²C⁺, ¹³C⁺, and H⁺ with the same velocity 120 keV/u were performed using the simulation code SIMNRA [1]. The intensity of the spectra was determined by fitting the measured spectra with the simulated spectra of ¹²C⁺ and ¹³C⁺ using the least-squares method. The post-acceleration proportions of ¹²CH⁺ and ¹³C⁺ in the mixed beam were determined to be 62.4% for ¹²CH⁺ and 37.6% for ¹³C⁺ for the present case. The validity of the obtained proportions can be confirmed by comparing the calculated H⁺ spectrum from the obtained intensity of ¹²CH⁺ ion with the measured H⁺ spectrum (Fig. 1).

The ${}^{13}C^+$ ratio obtained is higher than the estimated ratio of pre-accelerated ions. This is due to the chargeexchange process at the high-voltage terminal. At the terminal, the pre-accelerated negative ions collide with the charge stripper gas, producing positively charged ions through an electron loss process. The molecular ion could dissociate at this stage due to the collision process. In this sense, the fraction of molecular ions depends on the partial pressure of the charge stripper gas. As complementation, we measured the fraction of ¹³C⁺ in the accelerated m/q = 13 ion beam as a function of the partial pressure of the change stripper gas. The partial pressure was monitored by the vacuum pressure of the accelerator at the terminal. The results show that the ¹³C⁺ fraction in the beam m/q = 13 increases with the partial pressure of the charge stripper gas (Fig. 2) and reaches almost 100% for the high partial pressure of the stripper gas.

The results demonstrate that Rutherford backscattering can effectively quantify individual components in mixed molecular ion beams. This information is vital for characterizing the composition of the accelerated mixed beam and understanding the molecular ion acceleration process.



Fig. 1. Rutherford backscattering spectra of the accelerated m/q = 13 ion beam, alongside simulated spectra of ${}^{12}\text{CH}^+$ and ${}^{13}\text{C}^+$, utilized for determining post-acceleration proportions. Also shown is the sum of the simulated spectra ${}^{12}\text{CH}^+$ and ${}^{13}\text{C}^+$.



Fig. 2. The relationship between the vacuum pressure at the terminal of the accelerator, which monitors the partial pressure of the charge stripper gas, and the proportions of $^{12}CH^+$ and $^{13}C^+$.

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3.

ACCELERATOR MASS SPECTROMETRY



New Ta-covered wheel for Cl-36 AMS

3.1 Status report of the Tsukuba 6 MV multi-nuclide AMS system in FY 2023

K. Sasa, T. Takahashi, M. Matsumura, T. Yoshida, A. Sakaguchi, T. Matsunaka¹

The Tsukuba 6 MV multi-nuclide AMS system was operated for a total of 43 days and 516.6 hours in FY 2023. Figure 1 shows monthly-measured rare nuclides from April 2023 to March 2024. During the year, 595 samples were measured, mostly for ³⁶Cl and ¹²⁹I, and test measurements of ²¹⁰Pb were performed in the development of AMS. Table 1 shows the breakdown of the measured samples.



Fig. 1. Monthly-measured nuclides from April 2023 to March 2024.

Item	Reagent & Blank	Standard samples	Analyzed samples	Total
C1-36	8	4	7	19
I-129	63	42	407	512
Pb-210	55	6	3	64
Sub total	126	52	417	595

Table 1. Number of measured samples in FY 2023.

³⁶Cl-AMS

Cl⁻ extracted from AgCl was positively charged after passing through a carbon foil ($3.6 \mu g/cm^2$) at the acceleration voltage of 6.0 MV, and ${}^{36}Cl^{7+}$ was detected at 48 MeV in ${}^{36}Cl$ -AMS. There is a problem with isobar ${}^{36}S$ contaminations from the Cu wheel disk containing sample cathodes when Cl⁻ is extracted from the Cs sputtering ion source. We attempted to suppress isobar ${}^{36}S$ contaminations by covering the surface of the Cu wheel disk with a 0.50 mm thick Ta plate. As a result, the contribution of ${}^{36}S$ was reduced by about a factor of 50, as shown in Figure 2. In FY 2023, we have conducted joint research with Guangxi Normal University in China to measure anthropogenic ${}^{36}Cl$ in soil, and also with KEK to measure ${}^{36}Cl$ in activated chloride sheathing covering cables in the large accelerator room.

¹²⁹I-AMS

¹²⁹I-AMS has been performed under the beam condition of ¹²⁹I⁵⁺ with 30.0 MeV using an Ar gas stripper at the terminal voltage of 5 MV. In FY 2023, ¹²⁹I-AMS was mainly applied to ¹²⁹I tracer studies for ocean circulation studies, the discharge of ALPS-treated water to the sea at Fukushima and Iodine-129 deposition from atmosphere in the sample preparation rooms.

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Fig. 2. Comparison of the ratios of ³⁶S to ³⁵Cl counts between the Cu wheel (old) and the Ta-covered Cu wheel (New), showing the drastic reduction of ³⁶S contamination. Measured sample: KNCl-STD (³⁶Cl/Cl = 5.000×10^{-12}).

²¹⁰Pb-AMS

AMS development is underway for ²¹⁰Pb (a half-life of 22.2 years) in FY2023. PbO₂⁻ was successfully extracted as Pb negative molecular ions up to 550 nA from Pb₃O₄. Pb⁵⁺ was accelerated at the acceleration voltage of 3 MV, and a test detection was performed for a mass number of 210 using the gas ionization chamber. Detail of the ²¹⁰Pb AMS development is presented in Ref. [1].



Measurement history

Fig. 3. Cumulative number of measured samples for AMS since 2016.

Figure 3 shows the cumulative number of measured samples since the AMS system started operation. A total of 5,147 samples was measured from FY 2016 to 2023. AMS of the radioactive halogens ³⁶Cl and ¹²⁹I has been performed mainly at the Tsukuba 6 MV multi-nuclide AMS system since FY2020 [2].

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3.2 Riverine ¹²⁹I dynamics during high-flow events in the Abukuma and Niida rivers

Y. Wakiyama¹, M. Matsumura, T. Matsunaka², S. Hirao¹, K. Sasa

The long-lived ¹²⁹I (half-life: 15.7 million years) is one of radionuclides of interest released by the Fukushima Daiichi Nuclear Power Plant (FDNPP). Post-accidental research has revealed impact of ¹²⁹I release/deposition on various environmental media, such as rainwater, soil, and seawater. However, there have been few investigations on ¹²⁹I redistribution on terrestrial environment. To tackle this issue, we have analyzed river water samples collected from rivers under high-flow conditions since 2019 in the flamework of Environmental Radioactivity Research Network Center (ERAN).

The first set of samples were collected at the Kuroiwa site (catchment area 2890 km² and catchment mean ¹³⁷Cs inventory 103 kBq/m²) on midstream of the Abukuma river in July and October 2018, the second at the Haramachi site (206 km² and 853 kBq/m²) on downstream of the Niida river in August 2016 and October 2017, and at the Warabidaira site (29 km² and 1420 kBq/m²) on Hiso river, an upstream tributary of the Niida river, in August 2016. All river water samples were filtered with a 0.45 µm membrane filter, and thus obtained samples of suspended solids (SS) and filtrates were subject to pretreatment, ¹²⁹I/¹²⁷I ratio measurement by accelerator mass spectrometry, and ¹²⁷I concentration measurement by ICP-QQQ-MS [1].

Table 1 summarizes results of ¹²⁹I analyses. ¹²⁹I concentrations and ¹²⁹I/¹²⁷I ratios at the Kuroiwa site were in range of the background level before the FDNPP accident. Mean values of ¹²⁹I concentrations and ¹²⁹I/¹²⁷I ratios were the highest at the Warabidaira site, followed by the Haramachi site, and then the Kuroiwa site. This magnitude-relation coincided with that of catchment mean ¹³⁷Cs inventory, as an index of ¹²⁹I deposition level in the catchments. Apparent distribution coefficients also appeared to depend on the catchment mean ¹³⁷Cs inventory. These results suggest that Fukushima-derived ¹²⁹I may have a strong affinity to soil/sediment particles.

Site (river name)	Sample collection period	n	Mean dissolved ¹²⁹ I conc.	Mean ¹²⁹ I conc. in SS	Mean distribution coefficient	Mean ¹²⁹ I/ ¹²⁷ I in water	Mean ¹²⁹ I/ ¹²⁷ I in SS
			$(\mu Bq/L)$	(mBq/kg)	(L/kg)	(10^{-9})	(10^{-9})
Kuroiwa	5-7 Jul. 2018	5	0.179	1.03	4330	2.28	2.25
(Abukuma)	1-3 Oct. 2018	7	0.0677	0.437	6060	2.25	4.81
Haramachi	16-17 Aug. 2016	5*	0.174	4.07	22500	5.53	27.6
(Niida)	22-23 Oct. 2017	6	0.129	2.36	22200	4.92	31.3
Warabidaira (Hiso)	16-17 Aug. 2016	5	0.236	11.2	56100	10.4	48.5

Table 1. Summary of results of 129 I analyses. *n* indicates the number of analyzed samples.

* *n* for Mean ¹²⁹I conc. in SS, Mean distribution coefficient, and Mean ¹²⁹I/¹²⁷I in SS were four.

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3.3 Iodine-129 in seawater near the Fukushima Dai-ichi Nuclear Power Plant before and after the discharge of ALPS treated water

H. Miura¹, T. Matsunaka², K. Sasa, T. Takahashi, M. Matsumura

Anthropogenic ¹²⁹I is released into the ocean mainly from nuclear weapon tests, nuclear reprocessing facilities, and nuclear accidents including the Fukushima Dai-ichi Nuclear Power Plant (FDNPP) accident. Since August 2023, Tokyo Electric Power Company (TEPCO) has been releasing advanced liquid processing system (ALPS)-treated water, which contains trace amounts of ¹²⁹I, into the Pacific Ocean. This study aims to obtain the distribution of ¹²⁹I in seawater using AMS, which will help us in predicting concentration of ¹²⁹I by the model-based calculation for the radiation dose assessment. In this year, seawater collected before the discharge of the ALPS-treated water was analyzed.

Surface seawater was collected in the coast of Fukushima Prefecture by the research vessel Shinsei Maru (October 2022), and near the FDNPP by a fishing vessel (April and July 2023). Extraction and purification of I were carried out at Low Level Radioactivity Laboratory (LLRL), Kanazawa University. The ¹²⁹I/¹²⁷I ratio was measured using the AMS at University of Tsukuba. A terminal voltage of 5 MV and a charge state of 5+ were chosen. The measured ratios were normalized against the Purdue-1 reference material (¹²⁹I/¹²⁷I = 8.37×10^{-12}) obtained from Purdue University. I-127 alkaline solution was measured by ICP-MS. The ¹²⁹I/¹²⁷I ratio and ¹²⁹I in the seawater were calculated using ¹²⁹I/¹²⁷I ratio from AMS and ¹²⁷I concentration from ICP-MS.

Figure 1 shows the distribution of ¹²⁹I (Bq/L) in the coast of Fukushima Prefecture. The ¹²⁹I concentration has remained at the pre-accident level (10⁻⁸ Bq/L) at points excluding the NP0 near the nuclear power plant. On the other hand, the ¹²⁹I concentration at NP0 was about two orders of magnitude higher than those at the other points. Figure 2 shows the sampling sites and ¹²⁹I concentrations near the nuclear power plant. The ¹²⁹I concentration near the nuclear power plant was about one order of magnitude higher on average, with a large variability of about two orders of magnitude, suggesting continuous leakage of contaminated water from the FDNPP. The next purpose of this study is analyzing seawater collected after the discharge of ALPS-treated water to evaluate the impact of the water discharge.

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Fig. 1. ¹²⁹I concentration in the coast of Fukushima Prefecture.



Fig. 2. Sampling sites within 5 km from the FDNPP (Left). ¹²⁹I concentrations in seawater near the FDNPP (Right).

3.4 Distribution of I-129 in the land area after the Fukushima Daiichi Nuclear Power Plant accident

M. Matsumura, K. Sasa, T. Matsunaka¹, R. Tomita², T. Takahashi, H. Matsuzaki³, K. Sueki

A significant amount of radioactive material was released into the environment after the accident after the accident at the Fukushima Daiichi Nuclear Power Plant (FDNPP) on March 11, 2011, with atmospheric emissions commencing on March 12 and peaking during March 15–16 and 20–22 (TEPCO, 2012). Anthropogenic sources such as nuclear-fuel reprocessing plants had already increased the ¹²⁹I level in the environment above its natural background (¹²⁹I / ¹²⁷I = 1.5×10^{-12} ; e.g., [1]). Total amounts of radionuclides discharged into the atmosphere by the FDNPP accident was estimated to be 8.1 GBq of ¹²⁹I (T_{1/2}= 1.57×10^7 y) [2] and 120 – 200 PBq of ¹³¹I (T_{1/2}= 8.02 d) [3]. The long-lived ¹²⁹I can be used as a tracer to retrospectively infer the level of ¹³¹I. We got a result that the average of ¹²⁹I concentration was (2.74 ± 1.35) $\times 10^8$ atoms/g prior to the FDNPP accident as ¹²⁹I background data at Fukushima. After the FDNPP accident, the weighted average of ¹³¹I / ¹²⁹I ratios was estimated to be (4.02 ± 0.81) $\times 10^{-2}$ in 5-cm-thick surface soils of Fukushima area corrected to the time of earthquake [4].

Thirteen years have passed since the accident, but ¹²⁹I released into the environment still exist through repeated deposition and resuspension. We investigated the concentration of ¹²⁹I and isotopic ratio of ¹²⁹I / ¹²⁷I in river water in Fukushima Prefecture, at five points on the Niida River, once per year, from 2014 to 2020. The samples were analyzed using AMS at MALT, The University of Tokyo, until 2015 [5], and at

UTTAC since 2016 [6]. The analysis results are $[\times 10^8]^{5}$ shown in Fig. 1. The ¹²⁹I concentrations are in the range $(0.35-2.8) \times 10^8$ atoms L⁻¹, and the ¹²⁹I / ¹²⁷I ratio is $(0.51-4.6) \times 10^{-8}$. It appears that, as time passes, the ¹²⁹I concentration decreases quickly at first, then slowly converges to a constant value.

This work was supported in part by the KAKENHI under grant Nos. of 15H02340, 19H04252 and 22H02001



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3.5 The performance of iodine-129 AMS measurements at the University of Tsukuba (FY 2023)

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We have reported the performance of ¹²⁹I measurements by accelerator mass spectrometry (¹²⁹I AMS) [1] since 2017. In FY 2023, 512 samples of ¹²⁹I were measured by preparing the AgI targets, as shown in Fig. 1. Figure 2 shows the measured blank values of AgI precipitated from carrier-reagent, called old iodine, and the ¹²⁷I beam current measured with a Faraday cup (MFC02-2) before injection to the accelerator. The



average of the ¹²⁹I/¹²⁷I ratio with its standard deviation (1 σ) of old iodine is (2.5 ± 1.5) × 10⁻¹⁴, while the ¹²⁷I beam current tends to decrease year by year from the average current of 3.2 µA in FY2016–2017 to 0.71 µA in FY 2023. Correspondence between beam extraction conditions and beam current values of the ion source is shown in Fig. 3. There does not seem to be a causal relationship between the beam current and its reduction. Another possible cause of the beam current decrease is the alignment of the target position inside the ion source.

Fig. 1. Details of measured ¹²⁹I by the AMS system in fiscal 2023.



Fig. 2. Temporal variation of the ratio of ¹²⁹L/¹²⁷I of old- iodine blank values and ¹²⁷I beam current.

¹Kanazawa University



Fig. 3. Correspondence between beam extraction conditions and beam current values of the S-4 (MC-SNICS) ion source.

The measured ¹²⁹L/¹²⁷I values were normalized by using Standard Reference Material (SRM), Purdue-1 STD (Z94-0597) and Purdue-2 STD (Z94-0596) with an ¹²⁹L/¹²⁷I ratio of 8.378×10^{-12} and 6.540×10^{-11} [2] (the value was revised in 2014 [3]), which was provided by the Purdue Rare Isotope Measurement Laboratory (PRIME Lab) at Purdue University, USA. The average ¹²⁹L/¹²⁷I ratios with its standard deviation (1 σ) are (7.820 ± 0.734) × 10⁻¹² (9.4%) and (6.117 ± 0.204) × 10⁻¹¹ (3.3%) for Purdue-1 (run number 595) and Purdue-2 (run number 54), respectively in FY2023. The ratios of (average of measured values) / (nominal value) are 0.933 and 0.935 for Purdue-1 and Purdue-2, respectively, which are in good agreement.

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3.6 Challenges in measuring Pb-210 using accelerator mass spectrometry

H. Takahashi, K. Sasa, T. Takahashi, T. Yoshida, M. Matsumura, A. Sakaguchi

Lead-210 (²¹⁰Pb) is a U-series nuclide with a half-life of 22.4 years and has been used as a chronological tool for the Anthropocene environmental samples. Although some methods exist for measuring ²¹⁰Pb, there is currently no method for measuring a large number of environmental samples with an activity of a few mBq in a short period of time. To achieve rapid measurement of ²¹⁰Pb in environmental samples, this study aims to establish a method for measuring ²¹⁰Pb using accelerator mass spectrometry (AMS).

Focusing on corals as environmental samples, we investigated (i) Pb concentration and separation from coral samples, (ii) synthesis of inorganic Pb compounds for AMS and (iii) Pb isotope measurements by AMS. Chemical separation methods using the AG1-x8 anion exchange resin and the Pb-resin were optimised to remove nuclides and matrices that could interfere with measurements during AMS. Synthesis of Pb compounds (α PbO, β PbO, Pb₃O₄, Pb(NO₃)₂, PbCO₃ and PbF₂) was attempted to obtain stable solid inorganic salts from aqueous lead nitrate solution. Negative ion beam extraction experiments were carried

out on the synthesised Pb compounds using a Cs sputter ion source in order to find suitable Pb compounds and extraction conditions under which high and stable beam currents could be obtained. Furthermore, we confirmed the relationship among the acceleration voltage, the Pb positive ion charge state after electron stripping, and the Pb ion beam transmission efficiency, as shown in Fig. 1. Finally, the charged particles with mass number 210 were observed with an ionisation chamber using a sample with a known 210 Pb concentration (210 Pb / 208 Pb = 10⁻⁹, 10⁻¹⁰).

The Pb separation with the AG1-x8 was able to purify Pb with high chemical yields of more than 97%. The main matrix and interfering nuclides were also removed by more than 99%. On the other hand, the Pb-resin did not sufficiently elute Pb ($61.7 \pm 0.7\%$). In attempts to synthesise solid Pb compounds, mainly β PbO, Pb₃O₄, β PbO+Pb₃O₄(4:6), Pb(NO₃)₂, PbCO₃ and PbF₂ were finally synthesised.



Fig. 1. Relationship between the Pb ion charge state and transmission (%) in each acceleration voltage.

As for the AMS experiments, the highest negative ion beam current with species of ${}^{208}PbO_2^{-}$ can be induced from Pb₃O₄, but the most stable current beam can be obtained from β PbO + Pb₃O₄(4:6). It was also found that negative ion beam of ${}^{208}PbO_2^{-}$ could be efficiently converted into positive ion beams of Pb⁵⁺ at an acceleration voltage of 3 MV (Fig. 1). Finally, the correlation between the count rates and the concentration of 210 Pb was observed using the ionisation chamber.

3.7 Seawater circulation analysis in the Southern and Indian Oceans by cross-sectional I-129 observation

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The Southern Ocean is suitable for detecting water masses containing anthropogenic ¹²⁹I ($T_{1/2}$: 15.7 million years) tracer since ¹²⁹I in the Southern Ocean is 3–6 orders of magnitude lower than that in the Northern Hemisphere. This study aimed to clarify seawater circulation in the Southern Ocean by vertical cross-sectional observations of dissolved ¹²⁹I and water mass structure analysis from the Southern Ocean to the Indian Ocean, where few studies have been conducted.

Seawater was sampled horizontally and vertically at six sites in the Southern and Indian Oceans (maximum depth: 5,230 m) during the JAMSTEC MR19-04 voyage (December 2019–February 2020) and horizontally at 15 sites from Australia to Showa Station in Antarctica during the National Institute of Polar Research SR21 voyage (November 2021–March 2022). After adding 1 mg of ¹²⁷I carrier (Deepwater iodine, ¹²⁹I/¹²⁷I: 1×10^{-14}) per 500 ml of seawater, iodine was purified by solvent extraction, and silver iodide targets were prepared. The ¹²⁹I/¹²⁷I ratio of the target was measured using an accelerator mass spectrometer and

normalized using the S-Purdue reference material (129 I/ 127 I: 8.378 × 10⁻¹²).

In the Antarctic Circumpolar Current (ACC) region of the Southern Ocean, anthropogenic ¹²⁹I was rarely observed in the horizontal and vertical directions (maximum depth: 5,230 m) and was below the prenuclear activity level and AMS quantification limit (Fig. 1). Anthropogenic ¹²⁹I was observed in the surface layer from the northern area of the Southern Ocean to the Indian Ocean, and the relationship between the horizontal distribution of ¹²⁹I and water temperature, salinity, and ²²⁶Ra provided information on the boundary between the Indian Ocean surface waters and the ACC. The vertical distribution of ¹²⁹I in the Indian Ocean decreased with increasing depth from the surface. At the southernmost site in the South Ocean, two spike layers of ¹²⁹I above pre-nuclear activity level were identified at 201 m and 1,670 m water depths, and an upper ¹²⁹I spike was also observed in the isodensity layer at one site to the north.



Fig. 1. Surface distribution of ¹²⁹I in the Southern and Indian Oceans during 2019–2022.

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4.

MATERIALS RESEARCH



A concrete sample compressed into the 1 mm diameter drilled hole in an Al foil of 20 μ m thickness, prepared for analysis of contained bound water by T-ERDA. ---- Report 4.2

4.1 Characterization of AlInN/AlN/GaN structures by means of positron annihilation

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GaN transistors have been extensively studied because of their potential for high-power and RF-frequency devices. For GaN power devices based on AlGaN/GaN heterostructure, a two-dimensional electron gas is generated at the interface between the AlGaN layer and the GaN layer because of spontaneous and piezoelectric polarizations. The deposition of $Al_xGa_{1-x}N$ with $x \ge 0.3$ usually starts to degrade its crystalline quality because of a lattice mismatch between AlGaN and GaN. A lattice matched $Al_{0.83}In_{0.17}N/GaN$ heterostructure has received attention as a potential candidate for suppressing defect introduction near heterointerfaces [1]. However, the growth of a high crystal quality AlInN layer with a high Al mole fraction is a great challenge because of the difference in growth temperatures for AlN and InN. It was suggested that the device performance can be improved by inserting a thin AlN spacer layer into the interface between AlInN and GaN layers [2]. In the present study, we used a monoenergetic positron beam, scanning transmission electron microscope (STEM), and energy dispersive x-ray spectroscopy (EDX) to characterize the AlInN/AlN/GaN structure [3].

The sample structure investigated was AlInN/AlN/GaN fabricated on a Si substrate. AlInN(10 nm)/AlN structures with different thicknesses for the AlN layers (0.75, 1, 1.5, and 2 nm) were deposited using a metal organic chemical vapor deposition (MOVPE) technique. The Al mole fraction x of the Al_xIn_{1-x}N layer was determined to be 0.18 from its lattice parameters using high-resolution Rutherford backscattering spectrometry.

Figure 1 shows high-angle annular dark-field (HAADF) STEM images of the AlInN/GaN, AlInN/AlN(1 nm)/GaN, and AlInN/AlN(2 nm)/GaN samples, respectively. As shown in Fig. 1(b), a sharp interface image between the AlN layer and the GaN layer was obtained for the sample with the 1-nm-thick AlN layer, but this was not the case for the other samples [Figs. 1(a) and 1(c)]. The observed variation in the microstructure of the



Fig. 1. HAADF-STEM images of (a) AlInN/GaN, (b) AlInN/AlN(1 nm)/GaN, and (c) AlInN/AlN(2 nm)/GaN. Depth distributions of Ga, Al, and In in those samples are shown in (d), (e), and (f), respectively.

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heterointerfaces agrees with the relationship between the thickness of the AlN layer and the electron mobility [3], suggesting that the interface roughness is one of the causes decreasing the carrier mobility in the channel region. The depth distributions of the atomic concentrations of Al, In, and Ga were measured by EDX, and the results are shown in Figs. 1(d)–(f). For the AlInN/GaN sample [Fig. 1(d)], the observed asymmetry of the depth profiles of Ga, Al, and In near the AlInN/GaN interface suggests an enhanced diffusion of Ga into the AlInN layer. By comparing the results for the samples with and without the AlN layer, it can be concluded that the diffusion of Ga into the AlInN layer.

Vacancy-type defects in the samples were probed using the positron annihilation technique. Details of this



Fig. 2. S-E relationships for AlInN/GaN and AlInN/AlN/GaN. Thicknesses of the AlN layers are shown in the figure, where "0 nm" refers to AlInN/GaN. Solid curves are fits to the experimental data. The inset shows depth distributions of *S* obtained from fittings.

technique are given elsewhere [4]. Figure 2 shows S values as a function of incident positron energy E for the AlInN/GaN and AlInN/AlN/GaN samples. The dotted line shows the S value measured at E = 20 keV for the sample without the AlInN/AlN layers, and it corresponds to the annihilation of positrons in the GaN layer. In the S–E plots, shoulders are observed at $E \cong 2-3$ keV. Because the mean implantation depth of positrons with this energy region is 30–60 nm, the observed S value at this energy region is mainly associated with the annihilation of positrons implanted into the GaN layer.

The solid curves in Fig. 2 are fits to the experimental data. Here, the effect of the positron annihilation in the AlInN and AlN layers on the *S* value was neglected because of their thinness and the diffusion of positrons toward the surface. The obtained depth distributions of *S* are shown in the inset of Fig. 1. It can be seen that the damaged regions near the heterointerface extend up to 110-120 nm. The species of defects causing the increase in *S* can be identified as Ga-vacancy-type defects such as a Ga vacancy and/or their complexes with N vacancies or impurities (mainly carbon). The introduction of such vacancies can be attributed to the atomic diffusion from the GaN layer into the AlN and/or AlInN layers. Because the channel region of carriers is located inside these defect-rich regions (110–120 nm), the defects introduced by the deposition of the InAlN/AlN layer could act as scattering and/or trapping centers and cause degradation in the electron mobility.

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4.2 Hydrogen analysis of concrete samples by T-ERDA

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Concrete is an effective neutron shielding material used for a bioshield etc. in nuclear power plants; however, the radioactivities produced by nuclear reactions involving neutrons accumulate in concrete during long-term operation of reactors. Evaluation of the distribution of the radioactivities contribute to the cost estimation and planning for the decommissioning of aged nuclear power plants. In general, the overall distribution can be estimated by combining a Monte-Carlo code such as MCNP and a radioactivity calculation code such as ORIGEN-S. We have addressed to improve the accuracy for predicting the distribution [1,2].

Hydrogen, a constituent element of the water molecule, is the element that most strongly reduces the energy of neutrons. Moisture content in the concrete is difficult to be determined accurately with a thermogravimeter-differential thermal analyzer (TG-DTA) due to decomposition of carbonates and evaporation of volatile elements. Prompt γ -ray analysis (PGA) can accurately analyze the moisture content, although there is a time-consuming process to use the equipment. Alternatively, we focused on elastic recoil detection analysis with transmission layout (T-ERDA) to analyze the hydrogen in a powder sample.

Test samples of JG-3, one of the geochemical standard references of the Geological Survey of Japan (GSJ), were prepared by mechanical compression into a disc-shaped hole in Al foil, as shown in Fig. 1. The diameter of the hole is 1.2-2.0 mm and the thickness of the Al foil, which also equals to the thickness of the compressed concrete, is $20-40 \mu$ m. By measuring the weight of the concrete, the density of the compressed concrete was estimated to be 2.5-2.7 g/cc, which agrees reasonably with the nominal density of solid concrete 2.6 g/cc. Notably, the beam size of $8-9.5 \text{ MeV He}^{2+}$ was ~0.1 mm diameter which is much less than the size of the disc sample. The sample was underlaid with a Ni foil of 20 or 25 µm thickness to prevent the incident He from entering the particle detector.

Figure 2 shows the energy spectrum of recoil H for incidence of 8 MeV He²⁺ on a concrete sample of 40 μ m thickness. The recoil H at the concrete surface appears at 3.5 MeV indicated by a black arrow, according



Fig. 1. Setup of the concrete sample for analysis of H by T-ERDA.



Fig. 2. Energy spectrum of recoil H from the concrete sample of 40 μ m thickness, measured using 8 MeV He²⁺.

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to the estimate using the data base SRIM. The depth scale obtained also using SRIM is indicated by red arrows. The recoil H generated deeper than ~20 µm cannot be observed since its kinetic energy is too low to pass through the Ni foil. From the H yield integrated in the depth range 0–7.75 µm, corresponding to recoil by 8–7 MeV He, the moisture content *M* can be determined by using the database of recoil cross sections IBANDL in a similar manner reported previously [3]. The value of *M* thus obtained is (0.72 ± 0.07) %, where the ±0.07% uncertainty is due to the instability of the He²⁺ beam current. This value certainly reproduces $M=(0.9\pm0.1)$ % determined by PGA for the same concrete sample.

Figure 3 shows the hydrogen spectrum of the same concrete sample of 20 μ m thickness, measured using 9.5 MeV He²⁺. The energies corresponding to H recoil at the surface and backsurface of the concrete sample, estimated using SRIM, are shown by red arrows. In this case, the recoil H signals from the whole depth range can be seen, in contrast to the case of Fig. 2. The moisture content can be determined similarly from the H yield integrated over the whole depth. However, to our regret, only the rough result of *M*=0.27–0.76% has been obtained. The reason for such large uncertainty is the appreciable instability of the beam current of He²⁺ during the measurements. Stabilization of the He ion source is an urgent issue for advanced hydrogen analysis.

In Figs. 2 and 3, the background yield seen under the H yield is probably due to nuclear reaction products emitted from the sample. Indeed, the background yield sensitively depends on the beam position on the concrete surface, as shown in Fig. 4 for the typical two cases. Identification of the origin of the background is now under way to find a suitable energy range of He^{2+} for low-background measurements.



Fig. 3. Energy spectrum of recoil H from the concrete sample of 20 μ m thickness, measured using 9.5 MeV He²⁺.



Fig. 4. Typical high-background spectra measured for two different beam positions on the JG-3 sample of 20 μ m thickness, measured using 9.5 MeV He²⁺.

Although several issues still remain, for example, instability of the He ion source, we have successfully established T-ERDA to determine H content in a powder sample. We are now planning to measure moisture contents of various kinds of concrete samples.

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4.3 Mössbauer study of antiferromagnetic FeMnMo alloy

H. Tanimoto, Y. Maeda, E. Kita

Metals and alloys normally show a dilatation and a decrease in the modulus with increasing temperature. On the other hand, alloys with almost no changes in size or modulus are called "Invar" or "Elinvar" alloys. Ferromagnetic FeNi or FeNiCr systems are widely used for the Invar and Elinvar alloys. However, the temperature insensitivities of the size and modulus are diminished under external magnetic fields. Development of antiferromagnetic Invar and Elinvar alloys attracts great interests because the temperature insensitivities of the size and modulus are maintained under the external magnetic field [1]. Masumoto et al. [2] reported that the temperature changes in the size or modulus of antiferromagnetic FeMn alloys became considerably small by addition of 4d elements like Mo and Nb. But the effect of these additives on the properties are not well understood. In the present study, the magnetic property of the antiferromagnetic FeMnMo was investigated by Mössbauer spectroscopy in addition to the temperature change in the Young's modulus by the anelastic measurement.



Fig. 1. Mössbauer spectra of Fe-24.4wt%Mn-3.8 wt%Mo. The observed data (+) can be fitted with one antiferromagnetic 6-line (blue) and one nonmagnetic single line (blue) below 370 K (purple; sum of blue and red lines) whereas by one nonmagnetic single line at 410 K.

Thin sheets of FeMnMo alloy were cut from the ingots prepared by arc melting in high purity Ar atmosphere. The sheets were ground into a reed shape and used for the anelastic measurement. Thin foils of 30 μ m thick were prepared by cold-rolling and chemical etching of the thin sheets, and used for Mössbauer spectroscopy in transmission geometry. Fig. 1 shows the Mössbauer spectra of Fe-24.4wt%Mn-3.8wt%Mo, indicating the temperature insensitivity of the modulus between 230 and 320 K. The spectra below 370 K can be fitted with one 6-line (antiferromagnetic γ -phase with FCC structure) and one single line (paramagnetic ϵ -phase with HCP structure). The hyperfine field of the γ -phase (the splitting of the 6-line) decreases with the increase in the temperature and the spectrum at 410 K is explained by one single line. Néel temperature, T_N of the γ -phase estimated from the temperature change in Mössbauer spectra was about 382 K and similar values were found from the magnetization and anelastic measurements. The disappearance of magnetorestriction by the magnetic phase transformation from ferro- or antiferromagnetic to paramagnetic is attributed to the origin Invar or Elinvar properties but the insensitive temperature range

of the present Fe-24.4wt%Mn-3.8wt%Mo is much lower T_N . The lattice parameter of the γ -phase shows the increase with the increase in Mo or Mn concentrations but the effects of Mo on T_N or temperature dependence in modulus seem somewhat different from those of Mn. Further study is now in progress in order to clarify the roles of Mo and Mn on the Elinvar characteristics of FeMnMo alloys.

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4.4 Evaluation of reactively sputtered Fe oxide thin film growth with Mössbauer spectroscopy

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Iron oxides such as spinel ferrite have a long history as magnetic materials and have served as bulk basic materials for soft magnetic materials, magnetic recording media and permanent magnet materials. In recent years, applications have expanded to electronic devices such as spintronics, and thin film growth has become an important technology for fabrication. The previous research has shown that continuous growth from metallic α -Fe to Fe₂O₃ is possible [1] when depositing iron oxide by sputtering. The growth mode changes from metal mode to compound mode according to the amount of O₂ gas in Ar, and the products and film formation rate change. Furthermore, there is a hysteresis of

film formation speed, and film formation cannot be controlled solely by gas flow rate and input power.

For the purpose of film deposition control, we clarify the relationship among the intensity of plasma emission spectrum (PES), film deposition rate, and products. The ⁵⁷Fe Mössbauer effect was used to identify the product, and the emission spectrum during film formation was analyzed using principal component analysis to investigate the film formation control factors [2].

By using a metal iron target, O₂ gas was introduced as a reactive gas into Ar gas, and iron oxide thin films were deposited with RF magnetron sputtering. By using MgO single crystal as the substrate, 14 samples were fabricated while paying careful attention to the oxygen flow rate and



Fig. 1. Plasma emission spectrum in a wave-length range between 200 and 1000 nm during the sputtering deposition of Fe in Ar gas.



Fig. 2. Mössbauer spectra of Fe-O reactively sputtered films with low O_2 flow rates.

growth mode history. Plasma emission during film formation was recorded in the wavelength range between 200 nm and 1000 nm seen in Fig. 1. The film thicknesses were approximately 50 nm, and the growth rates

Relative emission

were calculated from the film thickness determined by the X-ray reflection method. ⁵⁷Fe conversion electron Mössbauer spectroscopy (CEMS) was carried out on these thin films to determine the valence of Fe atoms. For CEMS, a RiKon5 CEMS detector (WissEL product) was used with He (1 % of iso-butane) gas mixture (Q-gas). The thin films have areas from 100 mm² to 200 mm² and were set into the CEMS counter. The surfaces of the samples were connected electrically by a carbon tape to prevent charge-up of the samples.

As the flow rate was increased from zero, the products changed from α -Fe to FeO, Fe₃O₄, and γ -Fe₂O₃. Figure 2 shows the Mössbauer spectra in the low oxygen flow region. In particular, almost single phase FeO

was fabricated at the oxygen flow rate of 0.2 sccm. For the higher oxygen rate region, similar results as the previous work [2] were obtained and hysteric behaviors were also observed at the boundary region between Fe₃O₄, and γ -Fe₂O₃, around 0.9 sccm of oxygen flow rate. Figure 3 shows the film formation rate in the Fe₃O₄ and γ -Fe₂O₃ generation region. A hysteresis phenomenon was observed near the oxygen flow rate of 0.9 sccm (transition region between metal mode and compound mode).

In order to analyze the relation between growth conditions and the properties of produced thin films, PES data were first pre-analyzed. The emission peak intensity



Fig. 3. O_2 flow rates dependence of growth rates. Arrows show the order of film fabrication, and the hysteretic result is clearly shown.

over the entire observed wavelength range was used, therefore there are a large number of emission spectra, namely over 3000 channels. Principal component analysis was applied to reduce the dimensionality. After normalizing the emission spectrum as shown in Fig. 1 to the intensity per unit time, the intensity of low intensity peaks was set to zero. The analysis determined principal component axes (PC1, PC2), and we verified whether it was possible to predict the deposition rate and valence. The preliminary analysis revealed that the deposition rates cannot be expressed by PC1 only, but are almost expressed by PC1 and PC2. Further increase of dimension, PC3, was not effective, meaning that the deposition rates can be expressed in 2 dimensions, PC1 and PC2. In the valence evaluation, there were clear regions of composition in PC1 and PC2. The principal component analysis can drastically reduce the dimension of data channels from hundreds to 2. It is found that the principal component analysis has potential in controlling sputtering film fabrications.

Authors would like to express their thanks to Prof. K. Mibu of Nagoya Institute of Technology for his kind advises to CEMS experiments.

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4.5 Mössbauer study of an iron-based nanocrystalline alloy after annealing in magnetic fields with various temperatures

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An iron-based nanocrystalline alloy, which is obtained by annealing of an iron-based amorphous alloy, has been attracting attention as a soft-magnetic core material for power inductor and transformer because of their high saturation magnetic flux density B_s and low coercivity H_c . In order to improve the magnetic properties, it is necessary, for example, to decrease the coercivity while maintaining high B_s , and highly control the crystallization from amorphous to nanocrystalline structure.



Figure 1 shows schematic drawing of nano-structure of an iron-based nanocrystalline alloy. It consists of nano-sized α -Fe crystal grains and a residual amorphous region. Improved

Fig. 1. Schematic drawing of nano-structure of an iron-based nanocrystalline alloy.

number density in the nano-structure and suppression and homogenization of grain size are desirable for the α -Fe grains produced by crystallization. However, it is difficult to achieve such highly controlled crystallization by a simple heat-treated process. In this study, magnetic energy is examined as a new thermodynamic parameter that is effective in controlling crystallization of amorphous alloys by annealing in a magnetic field.

It has been reported that the crystallization processes of iron-based amorphous alloys are influenced by the applied magnetic field. In particular, it is known that the in-field annealing can accelerate nucleation and induce crystallization [1,2]. On the other hand, a magnetic field strength around 20 T is required to induce distinct crystallization, meaning that it is not possible to apply this method to any industrial process in practice [3]. We have been investigating a method of inducing crystallization by multi-step heat treatment in which the applied magnetic field strength can be suppressed during the in-field annealing.

The crystallization properties of amorphous alloys annealed at lower temperatures than the crystallization temperature have been investigated, but there are no apparent differences in the XRD profiles after low-temperature annealing. In contrast, thermal analysis revealed that the crystallization temperature decreased by about 5 K after low-temperature annealing. In order to investigate the microscopic changes in amorphous alloys caused by such low-temperature annealing, Mössbauer spectroscopy was applied to amorphous alloys annealed in both nonmagnetic and magnetic fields.

The sample used in this study was an untreated amorphous precursor of a nanocrystalline alloy known as NANOMET whose iron concentration is 85-86 at% [4]. The sample was prepared to be 10×10 mm in size from a thin ribbon of 60 mm in width and 17 µm in thickness. The crystallization temperature T_x evaluated

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by using a differential scanning calorimeter (DSC) was 658 K at a heating rate of 10 K/min.

The heat treatment was carried out using a cryogen-free superconducting magnet and an electric furnace for in-field heat treatment at High Field Laboratory for Superconducting Materials, Institute for Materials

Research, Tohoku University. The annealing temperature was chosen to be a sufficiently low temperature at which crystallization does not occur.

Mössbauer spectroscopy was performed at room temperature for sample pieces cut into 10×10 mm from the ribbon. Conventional constant acceleration mode was used for measurements with a velocity range of ±12 mm/s. The velocity was calibrated via α -Fe at room temperature.

Figure 2 shows spectra for different stages during the sample preparation. A spectrum for the sample in the as-quenched amorphous stage is displayed as (a), showing a typical spectrum of iron-based amorphous structure with an asymmetric pattern. No trace of iron oxides was detected in all samples. The spectrum does not change much after annealing sufficiently below the crystallization temperature, 592 K($0.9T_x$), at 0 T (see (b)). When the sample was annealed above the crystallization temperature 700 K (c), the spectrum shows a clear trace of α -Fe with a residual amorphous part in a crystallized sample.

To understand the small change in the structure of alloys with annealing under magnetic fields, an analysis proving distribution in hyperfine field will be effective.

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Fig. 2. Mössbauer spectra of (a) as-Q, (b) annealed at low temperature, and (c) crystallized samples.

4.6 Mössbauer study of superparamagnetic iron oxide nanoparticles

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Magnetic properties of nanoparticles (NPs) had been studied for a long time because these show drastic change owing to their size and have developed novel applications in addition to their conventional applications. For example, bio-medical application has been widely spread for such as a medical field of therapy and diagnostics, including magnetic NP hyperthermia, Magnetic Particle Imaging (MPI) and contrast agencies for magnetic resonance imaging (MRI). The contrast agency for MRI has already been in practical use and commercially available. To understand the specification for these applications, basic physical properties of MNPs must be carefully investigated.

Magnetic property of nano-particles is described by super-paramagnetism (SPM) due to a time dependent magnetization behavior against an external magnetic field, so called as relaxation phenomena. However, relaxation phenomena are quite different when the particle sizes are not mono-dispersed. Major relaxation mechanism of SPM particles is Néel mechanism in which thermal relaxation of magnetization in a small particle is responsible. The characteristic time of Néel mechanism τ_N is expressed as,

$$\tau_{\rm N} = \frac{\sqrt{\pi}}{\sqrt{\sigma}} \tau_0 \frac{1}{2} \exp(\sigma), \qquad (1)$$

where $\sigma = KV/(k_{\rm B}T)$ and $\tau_0 = 10^{-10}$ s [1]. Table 1 shows the calculated $\tau_{\rm N}$ based on eq. (1) providing $K = 10^4$ [Jm⁻³], T = 300 [K]. Here, $k_{\rm B} = 1.38 \times 10^{-23}$ [JK⁻¹]. It is clear that $\tau_{\rm N}$ is changed from an order of 10^{-24} s to 10^{-3} s when the volume of nano-particle increases by 8 times. This calculation supports large change in $\tau_{\rm N}$.

To investigate a frequency-dependent magnetic property, an AC magnetometry has been often utilized where the magnetization responds to an AC external magnetic field. The frequency of the AC magnetic field ranges from 1 Hz to 10 kHz for commercial susceptometers. For those, the corresponding relaxation time is from 1 s to 10^{-4} s. Mössbauer spectroscopy (MS) has a measuring time as fast as 10^{-6} s and information on relaxation at the time scale can be obtained and narrow range of measuring time can be covered by

Table 1. τ_N calculated for a cube with a side of a nm.

a [nm]	Volume [m ³]	$ au_{ m N}$ (s)
10	10 ⁻²⁴	6.4×10^{-24}
12.6	2 ×10 ⁻²⁴	5.0×10^{-9}
15.8	4 ×10 ⁻²⁴	4.5×10^{-7}
20	8 ×10 ⁻²⁴	1.9×10^{-3}



Fig. 1. TEM photo of nanoparticles.

changing the sample temperature. In order to make the relaxation behavior clear, we have studied a commercially available magnetic fluid (Resovist, product of Fujifilm RI Pharma) with MS measurements [2]. Resovist was a water-based suspension of superparamagnetic iron oxide (SPIO) NPs and the structure

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was not simple in order to stabilize their dispersity as suspension [3].

We choose the simpler structure of nanoparticle system than Resovist to avoid the influence of size distribution. The nanoparticle sample was provided by Future Materials Inc. TEM photo of the nanoparticles is shown in Fig. 1. The mean diameter of SPIO nanoparticles is 5.11 nm with a standard deviation of 1.37 nm. The toluene-based dispersion of the Fe₃O₄ nano particles and dried NPs were utilized for measurements. For the MS measurement, a sample holder made of a 2 mm thick Pb plate with a 10-mm diameter hole at the center was used, as shown in Fig. 2. The sample of 20 mg was placed into the hole and was immobilized by raising the temperature up to 70°C with mixing a paraffin wax of which the melting temperature is 60°C. The temperature in the measurements ranged between room temperature and 10 K by using a closed cycle refrigerator [1].

Mössbauer spectra recorded at temperatures from room temperature to 10 K are shown in Fig. 3. The peak intensity of 0.5% at room temperature is remarkably small compared with those of lower temperatures. The peak intensity was 3.6% at 250 K and reached 7% at 200 K. It was still small even at 250 K. This may be due to the fixing of the sample by the paraffin wax and the sample was not well immobilized near room temperature. Rigid support was established at 200 K. At 10 K, the spectrum was assigned to be that of maghemite. Between 30 K and 100 K, Mössbauer spectra showed patterns originated from a typically thermal-fluctuated magnetic state [3]. Compared with the spectra of Resovist, the temperature range with a spectrum showing a clear fluctuation is shifted to lower and the temperature dependence of MS has a similar trend. An



Fig. 2. The sample holder. The Pb sample plate has a hole of 10 mm in diameter for the sample space.



Fig. 3. Mössbauer spectra of SPIO NPs.

ideal spectrum from mono-dispersed particles cannot be detected and one with superposition of bimodal particle sizes is observed. From the result, the size distribution affects much on the magnetic relaxation even in the simple shaped sample seen in Fig. 1.

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4.7 Operando NRA on all-solid-state Li ion battery using ⁷Li isotope

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All-solid-state Li ion batteries, which can safely increase the capacity of Li-ion batteries, have attracted much attention. At present, the problem of insufficient conductivity remains for practical use. In order to understand the origin of this problem, we observed the behavior of Li in the battery using ⁷Li (p, α) ⁴He nuclear reaction analysis (NRA). Here, the natural abundance of Li isotopes is as follows: ⁷Li (92.4%), ⁶Li (7.6%), and we note that the above NRA observes only ⁷Li. So far, we have studied the all-solid-state Li ion battery as a sample, which has a simple structure but is practical enough. The sample consists of a stacked layer of Li (1.5 µm)/Li₃PO₄ (1.5 µm)/Au (100 nm) deposited on a Si₃N₄ membrane with 100 nm thickness, as shown in Fig. 1. The Si₃N₄ membrane is supported on a Si wafer. The Li layer, Li₃PO₄ layer, and Au layer correspond to the negative electrode, electrolyte, and positive electrode, respectively.

In the previous study, it was difficult to visualize the Li behavior at the interface between the negative electrode and the electrolyte, because both of these two layers contain Li [1]. In the present study, we prepared the electrolyte (Li₃PO₄) using the ⁶Li isotope, which is invisible in our NRA method. It was expected that the use of ⁶Li for electrolyte allows us to trace only the ⁷Li isotope, which departs from the negative electrode (pure Li layer) and migrates into the electrolyte. The results were, however, much different from our expectations.

NRA was performed using a 1.5 MeV H⁺ beam from 1 MV Tandetron at UTTAC. A schematic of the setup is shown in Fig.1. The Si surface detector (SSD) was set at $\theta_e = 150^\circ$ with respect to the incident beam direction. The emitted α due to the (p, α) reaction and backscattered H⁺ were detected by the SSD. The surface normal of sample is oriented at $\theta_{tilt} = 30^\circ$ from the beam axis, so that the escape angle of α with respect to the surface normal is $\phi_e = 60^\circ$. We note that the H⁺ beam was incident from the substrate (Si₃N₄ membrane) side following the previous study [1].



Fig. 1. Sample structure and experimental arrangement of NRA.

The procedure of NRA measurements was as follows. At first, the bias voltage of +5.0 V was applied to the positive electrode (the Au layer) of the as-deposited sample (battery) to make the battery as the charged

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state. Then the first NRA measurement was carried out (see the blue line in Fig. 2 (a)). Next, the sample was discharged by reducing the bias voltage down to +0.1 V. We took the second NRA profile on this stage (see the red line in Fig. 2 (a)). Thus, we compared the ⁷Li distribution before and after discharge of the battery. Similarly, the behavior of ⁷Li was observed when the battery was recharged by taking the NRA profile with sweeping the bias voltage from +0.1 V to +5.0 V.



Fig. 2. (a) The NRA profiles before (blue) and after (red) the discharge process of battery (sample).(b) The NRA profiles before (red) and after (blue) the recharge process of battery.

In the blue line in Fig. 2 (a), we notice that ⁷Li is distributed throughout the sample even for the asdeposited sample. This implies that the diffusion and/or intermixing of ⁷Li and ⁶Li in ⁶Li₃PO₄ occurs quickly even at the room temperature. Therefore, the expected sharp interface between the negative electrode (⁷Li layer) and the electrolyte (⁶Li₃PO₄) was not well observed. The interface is, however, still observed around the channel number of 2450 in Fig. 2 (a) and also in (b). From this fact, we deduce that the solid electrolyte consists of Li_x and Li_{3-x}PO₄. In other words, it is a combination of the mobile Li_x atoms (or ions) and the stiff framework of Li_{3-x}PO₄. The freely mobile Li atoms (or ions) in the electrolyte can be easily replaced by the Li atoms (or ions) from the negative electrode. Meanwhile, this phenomenon signifies that the retention of the electric power at the charged state with this combination of the negative electrode and the electrolyte is not easy at the room temperature.

Finally, we discuss on the results taken on the charged and discharged states of samples in short. With decreasing the bias voltage down to 0.1 V, the peak at the negative electrode disappears and the migration of ⁷Li into the positive electrode (Au layer) is seen. The recharge process is rather simple, i.e., the Au-Li alloy shows the partial decomposition and the considerable amount of Li returns to the negative electrode. The details of the whole movements of atoms, including P and O, must be clarified from the simultaneous analysis by Rutherford backscattering spectrometry (RBS) and NRA.

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4.8 Development of a rotational sample holder for ERDA on Fomblin oil surfaces

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For the purpose to keep and transport the ultra-cold neutron (UCN) more efficiently, the Fomblin oil coating on the walls of vacuum chambers and vacuum ducts attracts much attention [1]. Fomblin is a kind of fluorine-based oils, which does not stoichiometrically include hydrogen. While the Fomblin oil coating is expected to decrease the absorption cross section of walls for UCN, the direct measurements of hydrogen amount on the Fomblin oil surfaces have not been made in the past.

In order to determine the hydrogen concentration on the Fomblin oil surfaces, we developed a rotational sample holder for elastic recoil detection analysis (ERDA). In the design, a previously reported idea for high-resolution Rutherford backscattering spectroscopy (RBS) on an ionic liquid is adopted [2]. In the present study, we examined influence from the rotation (electric noises from the stepping motor and/or vibrational noises due to the looseness of mechanics) on the RBS and ERDA spectra.

Figure 1 shows the photograph of the developed rotational sample holder of 80 mm diameter. The wheel part is made of Al-based alloy (duralumin), and it is coated by the Au thin film with a thickness of 50 nm. The lower part of wheel is inserted into a small pool for oil. In the present study the measurements were carried out with leaving the pool empty. The simultaneous RBS and ERDA were performed with 2.5 MeV ⁴He²⁺ at the D-course in 1MV Tandetron at UTTAC. The arrangement of the sample holder with respect to the incident beam is shown in Fig. 2. Details of the detector setup are described in our previous paper [3]. For the measurement with the rotation, the rotational speed was set at 3 rpm.



Fig. 1. A photograph of the rotational sample holder.



Fig. 2. Setup for simultaneous RBS and ERDA. SSD stands for the B-implanted silicon surfacebarrier detector.

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Figure 3 (a) and (b) show the obtained RBS and ERDA spectra, respectively. The blue and red lines correspond to the measurements with and without the rotation of the sample holder, respectively. In the RBS spectra we can find the leading edge corresponding to Al (\sim 500 ch) and a peak of Au (\sim 900 ch). On the other hand, there are two peaks at \sim 700 ch and \sim 800 ch in the ERDA spectra. It is reasonable to assign them to the hydrogen at the interface between the duralumin substrate and the Au layer and the hydrogen on the surface of Au layer. We did not find any significant changes due to the rotation within the statistical errors.

As a next step, we will make the RBS and ERDA measurements on the Fomblin oil surfaces.



Fig. 3. (a) The RBS spectra taken on the rotational wheel with (blue) and without (red) rotation. (b) The ERDA spectra on the rotational wheel with (blue) and without (red) rotation.

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5.

ION-BEAM IRRADIATION EFFECT



Samples fixed on the A5-sized holder for ion beam irradiation experiments.

5.1 Fabrication and functional development of one-dimensional organic nanostructures using high-energy charged particles

M. Nobuoka¹, A. Taguchi¹, N. Miyake¹, W. Choi¹, S. Seki¹

Today's digital society has flourished to the point where it is no exaggeration to say that it is backed by the improvement of semiconductor microfabrication technology. While the miniaturization of semiconductors on the order of several nm, which is in the atomic domain, is just around the corner, it is said that the limitations of the fabrication method and material structure have begun to appear. In addition, in recent years, organic devices such as organic electroluminescence (EL) have already reached the stage of practical use, and they are expected to play a role in the next-generation semiconductor industry due to their versatile material selectivity. Therefore, the fabrication of organic nanomaterials to replace conventional inorganic materials and the establishment of fabrication technologies for such materials will be an immediate challenge, even in light of the recent SDGs.

The one-dimensional organic nanomaterials: nanowires fabricated by using energetic charged particles are based on the original technique that we have reported for a long time as SPNT (Single Particle Nanofabrication Technique) or STLiP (Single-particle Triggered Linear Polymerization) method (Fig. 1) [1,2]. The charged particles with kinetic energies of MeV-GeV travel through the material and form a cylindrical high-density activated region along their trajectory: ion track. In particular, excited and ionized states in ion tracks in organic materials, which are the starting points of chemical reactions, are densely formed and polymerization and/or cross-linking reactions between each organic

molecule proceed continuously. The insoluble gel-like one-dimensional structures formed in this way can be isolated as so-called organic nanowires, and we have reported two kinds of macrostructure in the nanowires obtained by different isolation methods [3] (Fig. 2). Vertically aligned nanowire array structures, in which flexible organic nanowires are freestanding on a support substrate, are expected to lead to the development of organic devices in multiple fields, which have not been



Fig. 1. Illustration of ion track in the organic materials, and SPNT and/or STLiP method.



Fig. 2. Schematic illustration of nanowire isolation and its morphology (wet and/or dry process) in STLiP method.

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feasible until now. In this project, we targeted various small organic molecules and polymers, to 1) create organic-inorganic hybrid nanomaterials, and 2) fabricate highly functional polymeric membranes.

Organic-inorganic hybrid nanowires

Polyhedral Oligomeric Silsesquioxanes (POSSs) are organic-inorganic hybrid molecules with organic side chains introduced into the Si-O framework, which exhibits solubility, thermal and thermo-mechanical properties, mechanical strength, optical permeability, gas permeability, dielectric constant, flame resistance, and many other physical properties. POSS films were deposited on silicon substrates by vacuum deposition and irradiated with Au beams (72-90 MeV) generated by the tandem accelerator at UTTAC. After irradiation,

the samples were heated at 200°C in a high vacuum to successfully isolate vertically aligned nanowires (Fig. 3). It was observed that the ease of nanowire formation varies depending on the different types of side chains. The nanowires are assumed to be approximately glasslike, and their application to optical waveguides and optical fibers is being investigated by taking advantage of their high dielectric properties and high light transmittance.



Fig. 3. Organic-inorganic hybrid nanowires. (a) molecular structure of POSS. (b) scanning electron microscope (SEM) tilted image of POSS-derived nanowires.

Ion beam modification of fluorinated polymers to incorporate new functions

Recently, nanochannels with densely fluorous interior surfaces were fabricated by supramolecular polymerization [4]. These nanochannels permeate water at an unprecedentedly high rate with a nearly perfect desalination, originating from the electrostatically negative fluorous interior surface, which can break water clusters to enhance the water permeability and can also provide a powerful electrostatic barrier for the incorporation of $C\Gamma$. We have attempted to fabricate membranes with a dense array of fluorous nanochannels in the same direction. Poly(vinylidene fluoride) (PVDF) films (KUREHA CORPORATION,

Mn²⁺

thickness: 28 μ m) were irradiated with Au beams (72-90 MeV) and then treated with XeF₂ exposure. By using electron spin resonance (ESR), it was confirmed that the radicals generated in the thin film by ion beam irradiation are eliminated by the XeF₂ treatment (Fig. 4). We plan to systematically evaluate whether the nano-channels that may have been formed in the thin films are highly fluorinated, and their water permeability.

$\int_{332}^{1} \int_{334}^{334} \int_{336}^{338} \int_{340}^{340} \int_{342}^{342}$ Magnetic field / mT Fig. 4. ESR spectra of irradiated PVDF films

Mn²⁺

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(upper) before and (lower) after exposing to XeF₂.

5.2 Morphology of the tracks of several-tens-MeV Au ions in YBa₂Cu₃O_{7-x} studied by cross-sectional transmission electron microscopy

H. Matsui¹, I. Yamaguchi¹

To accelerate the development of high-performance superconducting wires, vortex-pinning enhancement effects by ion irradiation have been extensively studied, particularly in candidate compounds, e.g., YBa₂Cu₃O_{7-x} (YBCO) and iron-based superconductors. Potential benefit of broken (segmented) columnar pinning centers (pins) has been a target of debate, and has provoked interest in low electronic energy loss (S_e) conditions (< 20 keV/nm in YBCO) where discontinuous or dotted 1D irradiation damage can be intentionally created instead of continuous 1D ones in the higher S_e regime. However, in YBCO, detailed morphology of such quasi 1D irradiation damage by low- S_e irradiation has not been well clarified by transmission electron microscopy (TEM). Regarding damage diameter (d) for example, the early evidence from TEM and critical-temperature (T_e) measurements suggested a constantly increasing dependence of d on S_e [i.e., $d(S_e)$] from $d \sim 1$ nm at $S_e = 13$ keV/nm up to $d \sim 10$ nm at $S_e = 40$ keV/nm (dashed line in Fig. 1a). However, recent cross-sectional TEM studies revealed 1D arrays of large spherical irradiation damage in $S_e < 20$ keV/nm. Furthermore, d in this case remarkably deviates from the conventional $d(S_e)$ trend (squares in Fig. 1a). To correctly conduct vortex pinning engineering in YBCO using low- S_e ions, it is essential to understand intrinsic $d(S_e)$ in the low- S_e region via detailed TEM evidence.

In this study [1], we performed high-resolution cross-sectional TEM measurements on YBCO thin films irradiated with 44-, 66-, and 84-MeV Au ions. The S_e (=11.6, 16.7, and 20.1 keV/nm) from SRIM code is near the onset of ion-track formation which was previously determined by plan-view TEM and T_c -measurements (about 13 keV/nm). YBCO epitaxial films on CeO₂-buffered SrTiO₃ substrates ($T_c = 90.1$ K)



◀ Fig. 1. (a) Electronic-energy-loss (S_e) dependence of iontrack diameter *d* in YBCO. Dashed line shows the general trend of open symbols that are early evidence from TEM and T_c measurements [Toulemonde et al., Nucl. Instrum. Methods Phys. Res. B 91 (1994) 108; Zhu *et al.*, Phys. Rev. B 48 (1993) 6436]. Blue squares are recent cross-sectional TEM data [Suvorova et al., J. Appl. Phys. 126 (2019) 145106]. The inset explains our definition of two "local" diameters of irradiation damage. (b) S_e dependence of maximum J_c at self-field and 77 K in our YBCO films obtained after Au irradiation in the dose range of 1×10⁹– 1×10¹² cm⁻². The dashed line indicates the upper-limit of asgrown samples. Numbers in parentheses are E_i of Au ions in MeV.

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were deposited by fluorine-free metal-organic deposition. The Au-ion irradiation was performed in a vacuum of 1×10^{-6} Torr at room temperature with a 6-MV tandem accelerator at the University of Tsukuba. The incident angle of ions was set normal to the film surface. The microstructure of the Au-irradiated YBCO films was evaluated using cross-sectional TEM images obtained with a JEOL JEM-2100F, at an acceleration voltage of 200 kV.

As shown in Fig. 2, we observed that the defect morphology changes from 1D arrays of spheres of about d = 5 to 10 nm in 44-MeV irradiation to their elongated variant (spheroid) statistically pierced by thin-lines of d = 1 to 2 nm and 3 to 5 nm in 66 and 84 MeV, respectively. This means that the thin-line damage appeared at the previous onset S_e of ion track formation, while the spherical damage appeared at S_e lower than the onset. We found that the "local" d values estimated within the thin-line segments in the tracks follow the conventional $d(S_e)$ trend (Fig. 1a), meaning that the trend holds over the entire S_e range, including the recently controversial $S_e < 20$ keV/nm, if we ignore the spheroid segments in the tracks.

We also found that the critical current densities (J_c) at self-field and 77 K in our YBCO films are enhanced by the Au irradiation at 55 MeV or higher, but not at 44 MeV or lower. This sharp variation in the width of irradiation-induced J_c with changing the incident ion energy (E_i) is correspondent well with the appearance of the thin-line damage (irradiation between 44- and 66-MeV).



Fig. 2. Cross-sectional TEM data of YBCO films irradiated with (a-d) 44-, (e-h) 66-, and (i-l) 84-MeV Au ions.

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5.3 Acquiring fundamental data for radiation protection of COTS-Grade FPGAs

H. Kukita¹, E. Miyazaki,² T. Shiobara¹, T. Kimura¹

The Japan Aerospace Exploration Agency (JAXA) and Nippon Dempa Kogyo Co., Ltd. (NDK) are currently conducting joint research and development of a flight model for demonstrations in LEO (Low Earth Orbit) [1].

For development of the initial prototype BBM (breadboard model), we selected a COTS (Commercial Off-The-Shelf) grade SoC-FPGA (System on a Chip Field-Programmable Gate Array). For the selected SoC-FPGA, a radiation resistance performance test was conducted in FY2022, thereby we confirmed that a function stop error is caused by SEU (Single Event Upset) but this error cannot be corrected by the ECC (Error-Correcting Code) function. We determined that ECC is insufficient for the space application of COT-grade SoC-FPGAs, hence implemented additional radiation countermeasures such as adding an SEM (Soft Error Mitigation) function and improving the scrubbing cycle. Then, we reconfirmed the soft error occurrence status. The beam irradiation tests were conducted using the UTTAC's 6MV tandem accelerator.

Test results (June 2023)

Beam irradiation tests with 12 MeV H⁺, 24 MeV Li³⁺, 36 MeV B⁵⁺, 42 MeV C⁶⁺, and 54 MeV O⁸⁺ were conducted.

Ion	Irradiation energy	Results
H^{+}	12 MeV	Error Free
Li ³⁺	24 MeV	Functional interrupt occurred with uncorrectable CRAM, BRAM and LUT errors
B^{5+}	36 MeV	Same as above
C ⁶⁺	42 MeV	Same as above
O ⁸⁺	54 MeV	Same as above

Note: Frequency of occurrence varies.

CRAM (Configuration RAM) : Memory area where circuit wiring information is set.

- BRAM (Block RAM) : Memory area used for storing user-designed programs, data, and parameter coefficients, etc.
- LUT (Look Up Table) : The part of the FPGA internal circuit that implements truth tables to realize combinational circuit functions, coefficient functions in signal processing circuits, etc., and are consisted with SRAM elements in some cases.

¹Nihon Dempa Kogyo Co., Ltd.

²Japan Aerospace Exploration Agency

Through detailed analysis of the above test results, we found that when two or more non-adjacent simultaneous errors occur, the error correction function does not work properly and the component stops functioning, leading to the SEFI (Single Event Functional Interrupt) condition.

To deal with this issue, it is necessary to take two countermeasures. One is for the architecture of the FPGA device itself such as CRAM, BRAM, SRAM circuit structure in the LUT section, flip-flops, and the other is for system soft error for the user logic section including the CPU. We are modifying our design. We plan to conduct another radiation evaluation after applying these countermeasures.

Reference

 Nihon Dempa Kogyo Co., Ltd. (NDK) and JAXA have started co-creation activities related to "Project to Create a New QCM sensor for Space Flight".

https://www.ndk.com/en/news/upload/NDK_JAXA_20240208_en.pdf https://global.jaxa.jp/press/2024/02/20240208-1 e.html 6. TECHNICAL REPORT



Improvement of the vacuum monitoring system at the high voltage terminal of the 6MV tandem accelerator.

6.1 Accelerator maintenance 2023

T. Moriguchi, T. Yoshida, Y. Yamato, S. Ishii, T. Takahashi, M. Matsumura, T. Nakazawa, K. Sasa

Recovery of the terminal vacuum gauge

The vacuum gauge installed at the high-voltage terminal in FY2022 [1] could not be operated around June 2023. During the regular maintenance with the tank opened in March 2024, the cause was found to be broken plastic optical fibers (POFs) caused by sparks (Fig. 1 (a)). As shown in Fig. 1 (b), we rewired the POFs to maintain the distance between them with conductive tape. To prevent spark damage to electronics in the terminal, band-shaped contacts were mounted inside the terminal shell (Fig. 1 (c)). After the regular maintenance inside of the tank, we confirmed operation of the terminal vacuum gauge (Fig. 1 (d)). Using the terminal vacuum gauge, we found that the terminal vacuum could potentially be used as a beam transport index because the terminal vacuum depends on the transport efficiency through the 6MV Pelletron tandem accelerator.



Fig. 1. (a) Broken plastic optical fibers (POFs) indicated by red allows. (b) Rewiring of POFs using conductive tape indicated by a red allow. (c) Band-shaped contacts mounted inside of the terminal shell.(d) The main display for monitoring of the terminal vacuum gauge.

Installation of new turbomolecular pump

We added an auxiliary turbomolecular pump (TMP) to the 03 line downstream of the 6MV Pelletron tandem accelerator (Fig. 2). Previously, with only one TMP operating from the terminal to the 03-1 valve, a number of inadequate vacuum evacuations occurred due to some 2-inch thin piping. For this reason, a new TMP with a displacement of 360 L/s manufactured by Leybold was added. This newly installed TMP improved the vacuum from 6.3×10^{-8} to 2.9×10^{-8} Torr.

Replacement of a hard disk of AccelNET control system

One RAID 1 hard disk failed on the AccelNET control system computer for the 6MV Pelletron tandem accelerator. There are five computers with RAID 1. At this issue, the alarm sounded from tsukuba1-10, which is placed at the end of the L4 line (AMS line) in the accelerator room. When an alarm sounds, the RAID disk is likely to fail. RAID 1 is a configuration that improves fault tolerance by writing the same data to two hard disks. All of the AccelNET control systems use RAID 1, but only tsukuba1-10 uses a 250 GB hard disk drive (HDD), while the others use 500 GB. We opened up tsukuba1-10 to see which hard disk had failed, removed the failed one, installed a new HDD, rebuilt it, and put it back together (Fig. 3).



Fig. 2. Newly installed TMP.



Fig. 3. Replaced RAID 1 hard disk tsukuba1-10.

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LIST OF PUBLICATIONS AND PRESENTATIONS

In larger method (%, 6%). For analysis of bydrogen in solids, possible experimental methods, in particular, of non-destructive type are extremely restricted because analysis techniques based on electronic matsitions of anoma are bardly agglicultile in this case. A notdifier and unique basers of elastic merid detection analysis (720A) using a last in beam is non-destructive detection of light another in a solid sample. The analysis of bydrogen in this case, it is solid sample. In this case, it is for antisension of the density agglicultile in this case, it is form hard collision of the density of the solid sample. In this case, it is form that collision of the density of the solid sample. In this destruction for antisension of the density of the solid sample. In this destruction for a first sample. The density of the solid sample is a mempanalyzed in the density of the solid sample is a solid frame of the density of the solid sample is a solid sample form hard collision of the density of the solid sample is a solid frame of the density of the solid sample is a solid sample is a frame of the density of the solid sample is a solid sample is a frame of the density of the solid sample is a solid sample is a frame of the density of the solid sample is a solid frame of the density of the solid sample is a solid sample (a, s), As density is a solid sample is a solid sample is a solid developed [10]. Laser field is a solid sample is a solid framework of the solid sample is a solid sample is a solid framework of the solid sample is a solid sample is a solid framework of the solid sample is a solid sample is a solid framework of the solid sample is a solid sample is a solid framework of the solid sample is a solid sample is a solid framework of the solid sample is a solid sample is a solid framework of the solid sample is a solid sample is a solid framework of the solid sample is a solid sample is a solid framework of the solid sample is a solid sample is a solid framework of the solid sample is a solid sample is a

Previously, we observed 30 distribution of bydrogen habities in a B-introduced Ai full by T-RELA. [195]. Autoally, such 30 analysis of hydrogen can be carried on by ading advantage of the enhanced recoil cross section as large a ~2 b sr⁻¹ due to the nuclear elastic memation of 8–010 MeV "He with H [16]. This allows sufficient count mar of menal it needed for the 3D observation by employing the collimated 10–100 pA He²⁺ beam of the µm size.

In our developing stage of nuclear-clustic T-ERDA, the main factors determining the spatial and depth resolutions typically in the μ m range were studied using test samples [17]. Further fundamental studies are needed to establish this method as a standard technique of hydrogen analysis. For this reason, we have carried out T-ERDA using samples of PPS (polyphenylene sulfide (C₆H₄S)_n) and natural muscovite. The former is a synthetic resin with well-known atomic composthose used in the previous work [17], which are abbreviated as PPS/AI(50), AI(10)/PPS/AI(50), and AI(20)/PPS/AI(50), where the numbers in the parentheses denote the thicknesses in µm. The thicknesses of the PPS film and the AI foils were determined by measuring the weight and area, assuming the demsitics of 2.70 and 1.35 g cm⁻³ for AI and PPS, respectively. The nominal purity of the AI foils is 99.99%. The measurement reproduced the nominal AI thicknesses of 10, 20, and 50 µm which is the error of $\pm 2\%$. The measured PPS thickness is 1.35 \pm 0.05 µm, which is thin enough to be used as a localized by marker. Furthermore, the PPS film was characterized by Reinferting backcattering spectroscopy (RBS) using 2 MeV the meaning in the composition analysis of the RBS specmenting in the composition analysis of the RBS specmenting in the composition analysis of the BBS specmenting backgroup in the bulk

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- 68. 細川律也,不破康裕,長谷川拓郎,広田克也,星野公邦,市川豪,猪野隆,岩下芳久,北口 雅暁,古賀淳,松崎俊,三島賢二,茂木駿紀,音野瑛俊, 關義親, 関場大一郎,嶋 達志, 清水春樹,清水裕彦,角直幸,角野浩史,谷田征輝,山下了,吉岡瑞樹,"J-PARC/BL05 に おける中性子寿命測定実験:高統計データを用いた信号抽出効率における系統誤差評価",日 本物理学会第78回年次大会,東北大学,仙台市(2023年9月16-19日).
- 69. 茂木駿紀,不破康裕,長谷川拓郎,広田克也,星野公邦,細川律也,市川豪,家城斉,猪野隆,岩下芳久,北口雅暁,牧瀬壮,松崎俊,三島賢二,森川滉己,長倉直樹,岡部宏紀,音野瑛俊,關義親,関場大一郎,嶋達志,清水春樹,清水裕彦,角直幸,角野浩史,谷田征輝,富田龍彦,上原英晃,山田崇人,山下了,矢野浩大,吉岡瑞樹,"J-PARC/BL05 における中性子寿命測定実験:2つの動作ガス圧力条件での中性子寿命解析",日本物理学会2024年春季大会,北海道大学,札幌市 (2024年3月18–21日).

7.4 UTTAC seminars

- 2024.1.9 Measurements of azimuthal anisotropy and quantum interference effects in $\sqrt{s_{NN}} = 19.6 \text{ GeV}$ Au+Au collisions at RHIC-STAR experiment, *Masataka Enomoto* (*University of Tsukuba*)
- 2024.1.9 Multiplicity dependence of W-jet production in pp collisions at $\sqrt{s} = 13$ TeV with LHC-ALICE, *Taiga Kawaguchi (University of Tsukuba)*
- 2024.1.9 Measurements of charged pions and (anti-)protons with respect to jet axis in pp and Pb-Pb collisions at $\sqrt{s_{NN}} = 5.02$ TeV at LHC-ALICE, *Koki Sato (University of Tsukuba)*
- 2024.1.9 Measurements of jet nuclear modification factor and azimuthal anisotropy in Pb-Pb collisions at $\sqrt{s_{NN}} = 5.02$ TeV with the LHC-ALICE to clarify the parton energy loss mechanism in Quark-Gluon Plasma and evaluate stopping power, *Takuya Kumaoka (University of Tsukuba)*
- 2024.1.17 Fabrication and characterization of a beam profile detector with scintillation fibers and MPPC for KEK PF-AR test beam line, *Yuki Asatani (University of Tsukuba)*
- 2024.2.28 Studies of nuclear spectroscopy with stopped and slowed-down RI beams and the developments of their related technologies, *Hideki Ueno (RIKEN Nishina Center)*

8.





Symbol of "achievement"

Doctor thesis

MUNDO, Rodrigo (Kanazawa University)	Study on fate of polycyclic aromatic hydrocarbons in Japan's coastal and marginal sea environment (日本の沿岸及び縁辺海域における多環芳香族炭化水素類の動態解明研究)
Master theses	
HONMA, Kouta	Degradation of carbon reinforced PFA studied by positron annihilation (陽電子消滅法を用いた炭素繊維強化 PFA の温度変化による劣化評価)
HIOKI, Ryu	Annealing properties of vacancy-type defects in GaN annealed by ultra-high pressure (Mg イオン注入 GaN の超高圧焼鈍による空孔型欠陥の焼鈍特性の研究)
TORII, Kazuma	Vacancy-type defects in AlInN/AlN/Ga structure studied by positron annihilation (陽電子消滅法による AlInN/AlN/Ga 構造中の空孔型欠陥評価)

Undergraduate theses

YANO, Yuichiro	Using Rutherford backscattering for elemental analysis of fast molecular ion composition (RBS を用いた高速分子イオンビームの成分分析)
MINAMI, Kenta	Development of Beam Current Monitor using Rutherford Backscattering (ラザフォード後方散乱を用いたビームカレントモニターの製作)
OKANO, Taiki	Development and Modification of FPGA based Radiation Measurement System (FPGA による放射線計測システムの開発及び改良)

9.

LIST OF PERSONNEL



High school students are touring UTTAC's (dark) back-side facilities, which are essential for operating the tandem accelerators.

Tandem Accelerator Complex

K. Sasa	Director, Associate Professor
D. Sekiba	Lecturer
T. Moriguchi	Assistant Professor
S. Ishii	Mechanical Engineer
T. Nakazawa	Mechanical Engineer
T. Takahashi	Electrical Engineer
Y. Yamato	Electrical Engineer
T. Yoshida	Electrical Engineer
M. Matsumura	Technical Staff
H. Muromachi	Administrative Staff

Research Members*

Division of Physics		
A. Ozawa	T. Moriguchi	K. Sasa
T. Yamaguchi (Saitama University [Cross appointment])		

Division of Applied Physics

E. Kita	D. Sekiba	S. Tomita	A. Uedono
H. Yanagihara	H. Tanimoto		

Division of Geoscience M. Kurosawa

Division of Chemistry A. Sakaguchi

Division of Information and Systems T. Kameda

Staff of Open Advanced Facilities InitiativeH. KudoH. Naramoto

M. Sataka

Graduate students

Graduate School of Science and Technology

^{*}**Research members** and **External users and collaborators** include the authors and coauthors within 3 years back from this fiscal year, as well as the members of research projects running at UTTAC.

T. Amagai	N. Kishi	Z. Shi	A. Yano
H. Takahashi	R. Hioki	K. Torii	K. Honma
K. Kawakami	M. Hiraga	U. Ozeki	K. Matsuda
T. Kanazawa	C. Kodaka	Y. Abe	T. Takahashi
H. Tanaka	R. Minami	H. Zhang	M. Tabata
Y. Maeda	K. Yamamoto		
Undergraduates			
R. Tanaka	Y. Kurihara	H. Yoshikado	G. Oya
K. Endo	T. Miyazaki	Y. Yano	T. Matsumoto
T. Okano	K. Minami	T. Abe	R. Tsuboi
E. Nishiyama	K. Yazaki		

External users and collaborators^{*}

T. Ishizuka	National Institute of Advanced Industrial Science and Technology (AIST)
S. Shiki	National Institute of Advanced Industrial Science and Technology (AIST)
H. Matsui	National Institute of Advanced Industrial Science and Technology (AIST)
T. Matsunaka	Kanazawa University
R. Mundo	Kanazawa University
S. Nagao	Kanazawa University
M. Ozawa	Kanazawa University
Y. Wakiyama	Fukushima University
H. Miura	Central Research Institute of Electric Power Industry
S. Seki	Kyoto University
M. Nobuoka	Kyoto University
W. Choi	Kyoto University
A. Taguchi	Kyoto University
N. Miyake	Kyoto University
S. Kobayashi	The University of Tokyo
G. Yoshida	High Energy Accelerator Research Organization (KEK)
H. Matsumura	High Energy Accelerator Research Organization (KEK)
E. Watanabe	High Energy Accelerator Research Organization (KEK)
M. Ishida	High Energy Accelerator Research Organization (KEK)
K. Tsugane	High Energy Accelerator Research Organization (KEK)
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S. Yamazaki	Japan Environment Research co., LTD.

K. Oishi	Japan Environment Research co., LTD.
N. Kinoshita	Shimizu Corporation
K. Sasaki	Saitama University
K. Okubo	Saitama University
Y. Koizumi	Saitama University
Y. Kikuchi	Saitama University
S. Nishizawa	Saitama University
K. Watanabe	Saitama University
N. Tomita	Saitama University
T. Suzuki	Saitama University
K. Ishiyama	Tohoku University
A. Iwase	Osaka Metropolitan University
H. Uno	SHI-ATEX Co., Ltd.
T. Shiobara	Nihon Dempa Kogyo Co., Ltd.
R. Onodera	National Institute of Technology, Ibaraki College